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(54) INORGANIC COMPOSITE POWDER AND COSMETIC COMPRISING THE SAME

(57) An inorganic composite powder has a satisfactory covering capability and a feeling of transparency, and a cosmetic blended the inorganic composite powder can cover such defects as blots and freckles on the human skin and insure a natural appearance.

The inorganic composite powder comprises two or more types of inorganic oxides having different refractive indexes respectively and sequentially laminated from the one with the highest refractive index at the bottom on a scaled substrate, wherein the difference in the refractive index between the utmost outer layer and adjoining inner layer is 0.6 or below, or the thickness of at least one of the second or higher inorganic oxide layers is within ± 20% of the value d given by the equation:

$$d = (\lambda \times X/4) / n$$

wherein λ indicates a wavelength of visual light, X indicates an odd integer, and n indicates a refractive index of the inorganic oxide.

In the cosmetic according to the present invention, the refractive index of the inorganic oxide use for forming the utmost outer layer of the inorganic composite powder is preferably 1.5 or below.

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$$d = (\lambda \times X/4) / n$$

wherein λ indicates a wavelength of visual light, X indicates an odd integer, and n indicates a refractive index of the inorganic oxide.

[0012] Said scaled substrate comprises preferably at least one of inorganic oxides selected from the group consisting of natural minerals such as mica, talc, and sericite, synthetic mica, synthetic sericite, plate-formed titanium oxide, plate-formed silica, plate-formed aluminum oxide, boron nitride, barium sulfate, and plate-formed titania and silica composite oxide.

[0013] The cosmetic according to the present invention is characterized in that said inorganic composite powder is blended therein and, the refractive index of the inorganic oxide use for forming the utmost outer layer of said inorganic composite powder is preferably 1.5 or below.

BEST MODE FOR CARRYING OUT THE INVENTION

[0014] Preferred embodiments of the present invent ion are described below.

[0015] A scaled substrate used in the present invention should preferably has an average particle diameter in the range from 2 to 20 μm and a thickness in the range from 0.05 to 1 μm. As the scaled substrate as described above, there can be enumerated such inorganic compounds as natural or synthetic mica, talc, sericite, and further plate-formed titanium oxide, plate-formed silica, plate-formed aluminum oxide, boron nitride, plate-formed barium sulfate, plate-formed titania silica composite oxide, but any other materials may be used so far as the materials have the size as described above.

[0016] The inorganic oxides which may be used for coating the scaled substrate and refractive indexes are shown below.

Titanium oxide (2.50)
Zinc oxide (2.0)
Zirconium oxide (2.2)
Cerium oxide (2.2)
Tin oxide (2.0)
Thallium oxide (2.1)
Barium titanate (2.4)
Aluminum oxide (1.73)
Magnesium oxide (1.77)
Yttrium oxide (1.92)
Silica (1.45)
Magnesium fluoride (1.38)
magnesium carbonate (1.5)
Calcium fluoride (1.43)

[0017] In addition, a mixture of these inorganic oxides and an inorganic oxide which is a composite or a solid solution may be used for this purpose.

[0018] The inorganic composite powder according to the present invention is formed by laminating thereon two or more inorganic oxides having different refractive indexes respectively selected from those listed above from the one having a higher refractive index into two or more layers. The inorganic oxide is selected based on the covering capability required for the inorganic composite powder to be laminated therewith. To obtain an inorganic composite powder with the higher covering capability, it is preferable to use an inorganic oxide with higher refractive index such as, for instance, titanium oxide for forming the first layer. To obtain an inorganic composite powder with the lower covering capability, it is preferable to use an inorganic oxide with the intermediate covering capability such as aluminum oxide for forming the first layer.

[0019] Then, the second and higher layers are formed for lamination, and to suppress reflection of light and obtain the feeling of transparency, it is necessary that the difference of a refractive index of the inorganic oxide used for forming the utmost external layer and that of an inorganic oxide used for forming the adjoining inner layer is less than 0.6. The difference between the refractive indexes should preferably be in the range from 0.05 to 0.5.

[0020] In this invention, to further suppress reflection of light and obtain the more excellent feeling of transparency, it is desired to limit a difference between refractive indexes of adjoining layers to 0.6 or below, and preferably to limit the difference in the range from 0.05 to 0.5. Namely, as the second layer it is desirable to select an inorganic oxide with the refractive index different by 0.6 or less from a refractive index of the inorganic oxide used form forming the first layer, and selection of inorganic oxides for the third or higher layers should be made based on the same principle. The smaller

ground can be hid while insuring the feeling of transparency. Therefore, an inorganic composite powder having the sufficient covering capability and the feeling of transparency can be obtained.

[0032] A cosmetic according to the present invention is described below.

[0033] Although the cosmetic according to the present invention comprises the inorganic composite powder according to the present invention as described above, the cosmetic gives the feeling of transparency and can hide defects on the ground, so that it can cover the human skin without giving any damage to the natural appearance.

[0034] As for the inorganic composite powder to be blended in the cosmetic, it is preferable to use an inorganic oxide with the refractive index of 1.5 or less, and more preferably in the range from 1.5 to 1.35 for forming the utmost outer layer of the inorganic composite powder. This configuration is preferable because the refractive index of oil blended in the cosmetic is less than 1.5 and the refractive indexes of the oil and inorganic oxide are close to each other with reflection of light suppressed. The most preferable inorganic oxide used for forming the utmost outer layer is silica.

[0035] A quantity of the inorganic composite powder blended in the cosmetic according to the present invention should preferably be in the range from 1 to 90 weight percent. When the percentage is less than 1 weight %, the excellent effect can not be obtained by blending the inorganic oxide in the cosmetic, and when the percentage is more than 90 weight %, such factors as the coloring performance and oily feeling originally required for cosmetics are lost.

[0036] A surface of the inorganic composite powder according to the present invention may be processed with silicone or any fluorine compound or the like when blended in a cosmetic.

[0037] The cosmetic according to the present invention includes at least one of various components included in ordinary cosmetics including, for instance, heavy aliphatic alcohol; heavy aliphatic acids; oils such as ester oil, paraffin oil, and wax; alcohol such as ethylalcohol, propyleneglycol, sorbitol, and glycerin; moisturizing agents such as mucopolysaccharides, collagens, PCA salt, and lactates; various types of nonion-based, cation-based, anion-based, or amphoteric surface surfactants; gums such as Arabian gum, xanthane gum, polyvinyl pyrrolidone, ethylcellulose, carboxylmethylcellulose, carboxyvinyl polymer, and denatured or not-denatured clay minerals; solvents such as ethylacetate, acetone, toluene; inorganic pigments/dyes; organic pigments/dyes; BHT; chelating agents; and perfumes. Also at least one or more of inorganic fillers such as silica, talc, kaolin, mica, physiological pigments, and various types of organic resins may be included.

[0038] The cosmetic according to the present invention can be manufactured in the normal way, and may be used in various forms such as powder form, cake-like form, pencil-like form, stick form, liquid form, and cream-like form. More specifically the cosmetic includes foundation, cream, emulsion, eye-shadow, nail enamel, eye liner, mascara, lip stick, pack, and cosmetic water.

[0039] The cosmetic according to the present invention provides the advantage that it is possible to obtain a cosmetic which can cover such defects as wrinkles, blots and melasma on human skin without losing the natural appearance.

[0040] The present invention is described in detail below with reference to the examples.

Example 1

[0041] 100 g of mica was added to and well dispersed in 1 liter of demineralized water, and further 56 g of titanyl sulfate aqueous solution corresponding to 20% titanium dioxide was added to the solution, and the solution was heated, agitated, and boiled for 5 hours. Then the solution was cooled down to the room temperature and filtered. The filterate was washed with water and dried under 110 ° C to obtain mica coated with titanium oxide hydrate. 100 g of mica obtained as described above was added to and well dispersed in 1 liter of demineralized water, and the solution was heated to 70 ° C. Then 111 g of zirconium ortho-sulfate aqueous solution corresponding to 10% zirconium oxide was gradually added to the solution keeping pH 5 with 5 weight % sodium hydroxide aqueous solution. After the addition was performed for about two hours, further 5 weight % sodium hydroxide aqueous solution was added to adjust pH 7 to pH 8, and then the solution was cooled down and filtered. The filterate was washed with water and dried under 110 ° C to obtain mica coated with titanium oxide hydrate and zirconium oxide hydrate.

[0042] 100 g of the mica obtained as described above was added to and well dispersed in 750 ml of demineralized water, and the solution was added to and mixed with a solution in which 11 g of aluminum chloride and 80 g of urea were dissolved in 250 ml of water, and the resultant mixture was heated to 90 ° C for 5 hours, cooled down to the room temperature, and then filtered. The filterate was washed with water and dried under 110 ° C, and heated under 600 °C for 5 hours to obtain mica coated with titanium oxide, zirconium oxide, and aluminum oxide in this order. Further 100 g of the mica obtained as described above was added to and well dispersed in 1 liter of a mixture of ethanol and water (mixing ratio: 7 vs 3) as a solvent. Then 250 g of ethanol solution containing ortho-ethyl silicate corresponding to 4 weight % silica was added, and the solution was heated to 50 ° C and kept at the temperature for about 10 hours, and then cooled and filtered. The filterate was washed with ethanol and washed sufficiently with demineralized water, and dried under 110 ° C to obtain the inorganic composite powder (A) sequentially coated with titanium oxide, zirconium oxide, aluminum oxide, and silica in this order.

sintered under 500 ° C for 3 hours to obtain mica coated sequentially with titanium oxide and zinc oxide in this order. The mica was sequentially coated with aluminum oxide and silica in the same procedure as that in Example 1 to obtain the inorganic composite powder (H) sequentially coated with titanium oxide, zinc oxide, aluminum oxide, and silica.

Example 9

[0051] Mica coated with zirconium oxide hydrate was obtained by following the same sequence as that in Example 1 excluding the point that the coating with titanium oxide was not performed. 100 g of the coated mica was added to and well dispersed in 1 liter of water and the solution was heated to 60°C. Then 370 g of magnesium chloride aqueous solution corresponding to 3 weight % magnesium oxide was added to the solution over 8 hours adjusting the pH 10 or more with 5 weight % sodium hydroxide aqueous solution, and then the solution was neutralized with diluted sulfuric acid to adjust the pH to 8, and then cooled to the temperature and filtered. The filterate was washed, dried under 110 ° C, and then sintered under 500 ° C for 2 hours. By coating the material obtained as described above with silica in the same way as that described in Example 1, the inorganic composite powder (I) sequentially coated with zirconium oxide, magnesium oxide, and silica was obtained.

Example 10

[0052] The mica coated with titanium oxide hydrate obtained in Example 1 was sintered for 3 hours under 800 ° C to obtain mica coated with titanium dioxide. The mica was observed with a scanning electron microscope with the magnitude of 1000 times to measure the size of 300 particles and the average size was obtained. The specific surface area of 2.5 m^2/g was calculated from the average particle size and the particle density.

[0053] By applying λ = 550, X = 1, and the refractive index of aluminum oxide n of 1.73 to the equation of $d = (\lambda \times X / 4) / n$, the value d of 79.5 nm is obtained. When the specific surface area and the density of the mica coated with titanium dioxide are taken into considerations, about 55 weight portions of aluminum oxide is required for 100 weight portions of mica coated with titanium dioxide for coating the aluminum oxide with the thickness of about 80 nm. Mica sequentially coated with titanium oxide and aluminum oxide was obtained by following the same sequence as that in Example 1 excluding the point that mica coated with titanium oxide was used in place of the mica coated with titanium oxide hydrate and zirconium oxide hydrate obtained in Example 1 and mixed with a solution in which 55 g of aluminum chloride and 400 g of urea were dissolved in 1250 ml of water. Further the mica sequentially coated with titanium oxide and aluminum oxide was coated with silica in the same way as that in Example 1 to obtain the inorganic composite powder (J) sequentially coated with titanium oxide, aluminum oxide, and silica.

Example 11

[0054] Mica coated with zirconium oxide hydrate was obtained by following the same sequence as that in Example 1 excluding the point that the coating with titanium oxide in Example 1 was not performed, and the mica was sintered for 3 hours under 800 ° C to obtain mica coated with zirconium oxide. The specific surface area was calculated in the same way as in Example 10, and the specific surface area was 2.7 m^2/g.

[0055] By applying λ = 550, X = 1, and the refractive index of silica n of 1.45 to the equation of $d = (\lambda \times X / 4) / n$, the value d of 94.8 nm is obtained. When the specific surface area and the density of the mica coated with titanium dioxide are taken into considerations, about 55 weight portions of silica oxide is required for 100 weight portions of mica coated with titanium dioxide to coat silica with the thickness of about 95 nm.

[0056] Silica was coated by following the same sequence as that in Embodiment 1 excluding the points that 100 g of mica coated with zirconium oxide was used in place of the mica sequentially coated with titanium oxide, zirconium oxide, arid aluminum oxide, and that 1325 g of ethanol solution containing ortho-ethyl silicate corresponding to 4 weight percent silica was added, and the inorganic composite powder (K) sequentially coated with zirconium oxide and silica was obtained.

Control 1

[0057] The mica (P) used in Example 1 was actually applied to and fully extended on a human skin and, the result of observation is shown in Table 1.

Control 2

[0058] The mica coated with titanium dioxide hydrate obtained in Example 1 was heated under 600 ° C for 5 hours to obtain the mica (Q) coated with titanium dioxide.

were compressed and molded.

[0065] The cake-shaped foundation was actually applied on a human skin, and the feeling of transparency was very high, yet wrinkles and pores on the skin were hardly seen, and the cosmetic film giving very natural feeling was provided.

Example 13

[0066] Emulsion-like foundation comprising the following compositions was prepared by using the inorganic composite powder (D) obtained in Example 4.

(1)	Demineralized water	63.6 (weight portions)
(2)	1,3-butylene glycol	6.5
(3)	Triethanol amine	1.5
(4)	Carboxymethyl cellulose	0.2
(5)	Bentonite	0.5
(6)	Inorganic composite powder (D)	6.5
(7)	Titanium oxide as pigment	1.5
(8)	Coloring pigment	Appropriately
(9)	Stearic acid	4.0
(10)	Monostearic acid propylene glycol	2.0
(11)	Cetostearil alcohol	0.2
(12)	Fluidized paraffin	3.0
(13)	Liquid lanolin	2.0
(14)	Myristic acid isopropyl	8.5
(15)	Methylparaben	Appropriately
(16)	Perfume	Appropriately

[0067] At first, the ingredients (2) to (8) were dispersed in demineralized water (1), and the solution was heated to 75 ° C. The ingredients (9) to (15) were fully mixed with each other under 80 ° C, and the mixture was homogeneously mixed in the dispersion above. The mixture was cooled and the ingredient (16) was added thereto to obtain the emulsion-like foundation.

[0068] The emulsion-like foundation was actually applied on a human skin, and the feeling of transparency was very high, yet wrinkles arid pores on the skin were hardly seen, and the cosmetic film giving very natural feeling was provided.

Table 1

Powder	Transparency	Covering capability	Total assessment
A	◎	◎	◎
B	○	◎	○
C	◎	◎	◎
D	◎	○	○
E	○	◎	○
F	◎	◎	◎

INTERNATIONAL SEARCH REPORT

International application No.
PCT/JP99/01502

A. CLASSIFICATION OF SUBJECT MATTER
Int.Cl[6] A61K7/02, 7/035, C01B13/14, 33/18, 21/068, 33/24, 33/42, 33/00, C01F7/02, 11/46, C01G23/04, C09C3/06
According to International Patent Classification (IPC) or to both national classification and IPC

B. FIELDS SEARCHED
Minimum documentation searched (classification system followed by classification symbols)
Int.Cl[6] A61K7/02, 7/035, C01B13/14, 33/18, 21/068, 33/24, 33/42, 33/00, C01F7/02, 11/46, C01G23/04, C09C3/06

Documentation searched other than minimum documentation to the extent that such documents are included in the fields searched

Electronic data base consulted during the international search (name of data base and, where practicable, search terms used)

C. DOCUMENTS CONSIDERED TO BE RELEVANT

Category*	Citation of document, with indication, where appropriate, of the relevant passages	Relevant to claim No.
X Y	JP, 5-230394, A (Kao Corp.), 7 September, 1993 (07. 09. 93), Full text (Family: none)	1-5 3, 4, 6
X Y	JP, 5-156174, A (Tayca Corp.), 22 June, 1993 (22. 06. 93), Full text (Family: none)	1, 2, 4, 5 3, 6
X Y	JP, 5-17329, A (Tayca Corp.), 26 January, 1993 (26. 01. 93), Full text (Family: none)	1, 2, 4, 5 3, 6
X Y	JP, 9-30935, A (Kao Corp.), 4 February, 1997 (04. 02. 97), Full text (Family: none)	1, 2, 5 3, 4, 6
Y	JP, 9-30917, A (Yuugen Kaisha Miyoshi Kasei), 4 February, 1997 (04. 02. 97), Page 4 (Family: none)	3, 6

[X] Further documents are listed in the continuation of Box C. [ ] See patent family annex.

* Special categories of cited documents:
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Date of the actual completion of the international search	Date of mailing of the international search report
18 June, 1999 (18. 06. 99)	29 June, 1999 (29. 06. 99)
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